JMYT-224US1 PATENT

#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant Ian William Carpenter et al.

Art Unit:

Application No:

To Be Assigned

Examiner

Filed:

Herewith

:

FOR:

CATALYTIC GENERATION OF

**HYDROGEN** 

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## **CONTINUATION OF:**

**Applicant** 

Ian William Carpenter et al.

Art Unit:

Application No:

09/646,497

Examiner

**Application Submitted:** 

September 18, 2000

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FOR:

CATALYTIC GENERATION

OF HYDROGEN

## PRELIMINARY AMENDMENT

Assistant Commissioner for Patents

Washington, D.C. 20231

SIR:

Prior to examination, please amend the above-identified application as follows.

#### IN THE SPECIFICATION:

Please insert the following paragraph at page 1, after the title:

This application is a continuation of U.S. Patent Application No. 09/646,497, filed on September 18, 2000, which was the National Stage of International Application No. PCT/GB99/00753, filed March 23, 1999.

Please replace the paragraph beginning at page 1, line 10, with the following:

A fuel cell works best when the anode is supplied with neat hydrogen. In the design of practical systems, however, other factors also need to be considered, including the availability, cost, supply, distribution, storage and release of clean hydrogen. When all these factors are taken into consideration, alternative methods of fueling can shown an overall advantage.

Please replace the paragraph beginning at page 1, line 16, with the following:

The issue of fueling is very dependent on the type of application. For example, the design of fuel cell powered passenger vehicles requires a compact and responsive supply of hydrogen which must provide comparable driving performance to that of a combustion powered vehicle, as well as achieving higher efficiency and improved emission standards. Although conventional and novel onboard hydrogen storage options are being developed, these do not seem likely to meet the target requirements for mass, size and cost, in time to be used for the first generation of fuel cell vehicles. Instead, the technology most likely to be implemented in the short term is the on-board generation of hydrogen from a liquid or liquefied fuel. On the other hand, the design of domestic systems for generating heat and fuel cell power is less constrained by the need for compactness and speed of response. Furthermore, as the most widely available domestic fuel is natural gas, the efficient conversion of methane to hydrogen is seen as a key development target.

Please replace the paragraph beginning at page 2, line 25, with the following:

However, the disadvantages of methanol are equally familiar, notably:

- (i) relatively high toxicity;
- (ii) high affinity for water, resulting in corrosiveness;
- (iii) absence of infrastructure for supplying vehicle fueling stations; and
- (iv) unsuitability for domestic use.

Please replace the paragraph beginning at page 3, line 1, with the following:

The question of supplying and distribution, in particular, has emerged as one of the key issues in the debate on the fueling of fuel-cell systems, with a strong case being made for the use of the most widely available fuels. This caused us to further investigate the feasibility of generating hydrogen from hydrocarbon fuels by the self-sustaining reaction of air and steam as can be accomplished *inter alia* by our HotSpot reactor.

#### IN THE CLAIMS:

Please replace claims 2, 3, 6-8, 10, 13, 18, 19, 21, and 22, with the following amended claims:

- 1 2. (Amended) A process according to claim 1 wherein the
- 2 stream is combined with the hydrocarbon and the oxygen-containing gas to form the
- 3 mixture after the self-sustaining partial oxidation of the hydrocarbon has
- 4 commenced.

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- 3. (Amended) A process according to claim 1 wherein the 1 2 hydrocarbon is a straight chain hydrocarbon or a branch chain hydrocarbon.
- 6. 1 (Amended) A process according to claim 1 wherein the
- hydrocarbon is selected from methane, propane, butane, hexane, heptane, normal-2
- 3 octane, iso-octane, naphthas, liquified petroleum gas, reformulated petrol and
- diesel-type fuels. 4
- 7. 1 (Amended) A process according to claim 1 wherein the 2 oxygen-containing gas is air.
- 1 8. (Amended) A process according to claim 1 wherein rhodium comprises 0.1 weight per cent to 5 weight per cent of the total weight of the 2 supported catalyst. 3
  - 10. (Amended) A process according to claim 1 wherein the refractory oxide support material is a mixture of ceria and zirconia.
  - 13. (Amended) A process according to claim 1 wherein the catalyst is pre-heated to a temperature at which self-sustaining partial oxidation of the hydrocarbon commences.
  - 18. (Amended) A process according to claim 1 wherein the mixture of the hydrocarbon and the oxygen-containing gas is fed to the catalyst when the catalyst has reached the temperature at which self-sustaining partial oxidation of the hydrocarbon will occur.
- 1 19. (Amended) A process as claimed in claim 1 operated in combination with a catalysed water-gas shift reaction for the reduction of carbon 2 monoxide in the hydrogen produced from the hydrogen.
- 1 21. (Amended) A process according to claim 19 wherein the water-gas shift reaction catalyst is added to the rhodium based catalyst for the 2 3 hydrogen generation reaction.

1 22. (Amended) The use in a fuel cell system of the process as 2 claimed in claim 1 for the catalytic generation of hydrogen.

# IN THE ABSTRACT:

Please add the abstract, which is attached as a separate sheet, to the application.

Respectfully submitted,

Christopher R. Lewis, Reg. No. 36,201 Attorney for Applicants

CRL/lrb

Dated: April 11, 2001

Suite 301 One Westlakes, Berwyn P.O. Box 980 Valley Forge, PA 19482-0980 (610) 407-0700

The Assistant Commissioner for Patents is hereby authorized to charge payment to Deposit Account No. **18-0350** of any fees associated with this communication.

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I hereby certify that this paper and fee are being deposited, under 37 C.F.R. § 1.10, and with sufficient postage, using the "Express Mail Post Office to Addressee" service of the United States Postal Service on the date indicated above and that the deposit is addressed to the Assistant Commissioner for Patents, Washington, DC 20231.

Kathleen Libby

#### ABSTRACT

A process for the catalytic generation of hydrogen by the self-sustaining combination of partial oxidation and steam reforming of a hydrocarbon comprises containing a mixture of the hydrocarbon, an oxygen-containing gas and steam with a catalyst comprising rhodium dispersed on a refractory oxide support material which is a mixture of ceria and zirconia. The hydrocarbons are straight chain or branch chain hydrocarbons having 1 to 15 carbon atoms and include methane, propane, butane, hexane, heptane, normal-octane, iso-octane, naphthas, liquefied petroleum gas and reformulated gasoline petrol and diesel fuels. The hydrogen generation process can be started by feeding the hydrocarbon and air to initiate partial oxidation, before steam is added. The hydrogen generation process can be started by feeding the hydrocarbon and air to initiate partial oxidation, before steam is added. The hydrogen generation process also may be operated in combination with a water-gas shift reaction for the reduction of carbon monoxide in the hydrogen generated.

#### VERSION WITH MARKINGS TO SHOW CHANGES MADE

#### IN THE SPECIFICATION:

Specification at page 1, after the title:

This application is a continuation of U.S. Patent Application No. 09/646,497, filed on September 18, 2000, which was the National Stage of International Application No. PCT/GB99/00753, filed March 23, 1999.

Specification at page 1, line 10:

A fuel cell works best when the anode is supplied with neat hydrogen. In the design of practical systems, however, other factors also need to be considered, including the availability, cost, supply, distribution, storage and release of clean hydrogen. When all these factors are taken into consideration, alternative methods of [fuelling] fueling can shown an overall advantage.

Specification at page 1, line 16:

The issue of [fuelling] <u>fueling</u> is very dependent on the type of application. For example, the design of fuel cell powered passenger vehicles requires a compact and responsive supply of hydrogen which must provide comparable driving performance to that of a combustion powered vehicle, as well as achieving higher efficiency and improved emission standards. Although conventional and novel on-board hydrogen storage options are being developed, these do not seem likely to meet the target requirements for mass, size and cost, in time to be used for the first generation of fuel cell vehicles. Instead, the technology most likely to be implemented in the short term is the on-board generation of hydrogen from a liquid or liquefied fuel. On the other hand, the design of domestic systems for generating heat and fuel cell power is less constrained by the need for compactness and speed of response. Furthermore, as the most widely available domestic fuel is natural gas, the efficient conversion of methane to hydrogen is seen as a key development target.

Specification at page 2, line 25:

However, the disadvantages of methanol are equally familiar, notably:

- (i) relatively high toxicity;
- (ii) high affinity for water, resulting in corrosiveness;
- (iii) absence of infrastructure for supplying vehicle [fuelling] fueling stations; and
- (iv) unsuitability for domestic use.

Specification at page 3, line 1:

The question of supplying and distribution, in particular, has emerged as one of the key issues in the debate on the [fuelling] <u>fueling</u> of fuel-cell systems, with a strong case being made for the use of the most widely available fuels. This caused us to further investigate the feasibility of generating hydrogen from hydrocarbon fuels by the self-sustaining reaction of air and steam as can be accomplished *inter alia* by our HotSpot reactor.

## IN THE CLAIMS:

- 2. (Amended) A process according to claim 1 wherein the
  stream is [introduced into the mixture of] combined with the hydrocarbon and the
  oxygen-containing gas to form the mixture after the self-sustaining partial oxidation
  of the hydrocarbon has commenced.
- 3. (Amended) A process according to claim 1 [or 2] wherein the hydrocarbon is a straight chain hydrocarbon or a branch chain hydrocarbon.
- 6. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the hydrocarbon is selected from methane, propane, butane, hexane, heptane, normal-octane, iso-octane, naphthas, liquified petroleum

4 gas, reformulated petrol and diesel-type fuels.

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- 7. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the oxygen-containing gas is air.
- 8. (Amended) A process according to [any one of the preceding claims] claim 1 wherein rhodium comprises 0.1 weight per cent to 5 weight per cent of the total weight of the supported catalyst.
- 1 10. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the refractory oxide support material is a mixture of ceria and zirconia.
- 1 13. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the catalyst is pre-heated to a temperature at which self-sustaining partial oxidation of the hydrocarbon commences.
  - 18. (Amended) A process according to [any one of the preceding claims] <u>claim 1</u> wherein the mixture of <u>the</u> hydrocarbon and <u>the</u> oxygen-containing gas is fed to the catalyst when the catalyst has reached the temperature at which self-sustaining partial oxidation of the hydrocarbon will occur.
  - 19. (Amended) A process as claimed in [any one of the preceding claims] <u>claim 1</u> operated in combination with a catalysed water-gas shift reaction for the reduction of carbon monoxide in the hydrogen produced from the hydrogen.
- 1 21. (Amended) A process according to claim 19 [or 20] wherein 2 the water-gas shift reaction catalyst is added to the rhodium based catalyst for the 3 hydrogen generation reaction.
- 1 22. (Amended) The use in a fuel cell system of the process as 2 claimed in [any one of the claims] <u>claim</u> 1 [to 21] for the catalytic generation of 3 hydrogen.

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# IN THE ABSTRACT:

The following abstract has been added:

# **ABSTRACT**

A process for the catalytic generation of hydrogen by the self-sustaining combination of partial oxidation and steam reforming of a hydrocarbon comprises containing a mixture of the hydrocarbon, an oxygen-containing gas and steam with a catalyst comprising rhodium dispersed on a refractory oxide support material which is a mixture of ceria and zirconia. The hydrocarbons are straight chain or branch chain hydrocarbons having 1 to 15 carbon atoms and include methane, propane, butane, hexane, heptane, normal-octane, iso-octane, naphthas, liquefied petroleum gas and reformulated gasoline petrol and diesel fuels. The hydrogen generation process can be started by feeding the hydrocarbon and air to initiate partial oxidation, before steam is added. The hydrogen generation process can be started by feeding the hydrocarbon and air to initiate partial oxidation, before steam is added. The hydrogen generation process also may be operated in combination with a water-gas shift reaction for the reduction of carbon monoxide in the hydrogen generated.